

NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

TECHNICAL NOTE 3887

EFFECT OF CONCENTRATION ON IGNITION DELAYS FOR
VARIOUS FUEL-OXYGEN-NITROGEN MIXTURES AT
ELEVATED TEMPERATURES

By E. Anagnostou, R. S. Brokaw, and J. N. Butler

Lewis Flight Propulsion Laboratory
Cleveland, Ohio

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December 1956

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EFFECT OF CONCENTRATION ON IGNITION DELAYS FOR VARIOUS
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SUMMARY

Ignition delays for mixtures of ethane, n-butane, isobutane, hydrogen, or propane with oxygen and nitrogen were measured using a flow system with two experimental procedures. Delays were measured in the temperature ranges 571° to 733° C for ethane, 444° to 715° C for n-butane, 564° to 713° C for isobutane, 594° to 652° C for hydrogen, and 540° to 681° C for propane. The effect of fuel concentration was measured for all fuels, and delays were inversely proportional to fuel concentration to the 0.6 to 1.7 power. The effect of oxygen concentration was measured for propane and the butanes, and, in general, delays decreased slightly with increasing oxygen concentration. However, at very low temperatures, for n-butane, the dependence on oxygen concentration increased. The two procedures gave different absolute values for the delays, but the trends were the same by either method. From the variation of delay with temperature at constant fuel and oxygen concentration, apparent activation energies were calculated.

INTRODUCTION

Spontaneous ignition temperature and spontaneous ignition delay have been of interest for many years, although results of different researchers are not usually comparable because of varying experimental conditions. In spontaneous ignition delay measurements, different techniques have given widely different results on the effect of a single variable. For example, in the work of references 1 and 2, increasing the fuel concentration did not affect the ignition delay, whereas reference 3 reported it resulted in shorter delays. To help elucidate the fuel concentration effect and to learn something about the ignition reactions taking place, work was begun at the NACA Lewis laboratory to measure some of these effects.

The system used was that of reference 4 in which preheated fuel and air were mixed quickly and then allowed to flow through a tube kept at constant temperature. This flow system was chosen since it

allowed the control of temperature and composition from the time the mixture was prepared until it ignited. Fuel was either bypassed continuously around the tube between runs while air flowed through the tube or the fuel and air were mixed and then bypassed for a short while before each run. In each case, fuel and air were preheated separately. Also, both techniques provided an air-mixture interface which contained all mixtures leaner than the one being studied. However, previous work (ref. 4) using nitrogen as the gas leading the fuel-air mixture into the tube showed that the delays measured with the air lead were really characteristic of the mixture and not of the interface.

This system, using only the procedure of bypassing fuel continuously between runs, was used to obtain the data on propane reported in reference 5. In that study ignition delay varied inversely with the propane concentration to some power n , where n ranged from 0.74 to 1.00, and with approximately the $1/4$ power of the oxygen concentration. Also, delays decreased with increasing temperature and pressure. These results were explained at least qualitatively by either slow reaction kinetics, which correctly described the fuel concentration effect, or by a surface reaction which described the fuel and oxygen effect.

The present work has extended the research to include other hydrocarbon fuels and hydrogen. It also includes further study of propane using a modified technique (fuel-air bypassing) by which it was hoped the effect of fuel cracking on the ignition delay could be determined. The hydrocarbons tested were ethane, isobutane, and *n*-butane; delays are reported in the temperature ranges 571° to 733° C for ethane, 444° to 715° C for *n*-butane, 564° to 713° C for isobutane, and 594° to 652° C for hydrogen. In addition, the ignition delays for propane measured with the modified technique and those remeasured using the original technique (fuel bypassing) are reported in the temperature range 540° to 681° C. The effect of oxygen concentration and temperature on the delays for the butanes and propane were also measured. No attempt is made to interpret these results using kinetics, although the discussion of reference 5 on this point could be applied.

APPARATUS AND PROCEDURE

The apparatus used in this research was essentially the same as the one of reference 4 mentioned previously and is shown in figure 1. Several slight modifications were made in order to use the method of bypassing the fuel and air together.

The ignition tube was a 3-foot-long, 50-millimeter-diameter Vycor tube surrounded by a metal jacket. The tube was heated mainly by air supplied from a 25-kilowatt heater. This air flowed through another surrounding insulated jacket. Temperature gradients in the tube, measured

axially by a movable thermocouple, were eliminated by six independently controlled heaters. The fuel and air were separately metered through critical flow orifices, preheated, and then mixed in a heated tangential jet mixer which had a volume of approximately 1 cubic centimeter. The residence time in the mixer ranged from 0.04 to 2 percent of the ignition delay.

In the original apparatus, there was a line for leading nitrogen instead of air. Since no more work with nitrogen was contemplated, this line (approx. 5/32" I.D.) was converted into an exhaust line for bypassing the fuel and air mixture. This is the line marked "Lower exhaust" in figure 2. A two-way solenoid valve (S) replaced the three-way one previously on this line and the electrical circuit was arranged so that when S and the fuel solenoid valve (F) were open, the fuel-air mixture would be exhausted without going through the ignition tube (fig. 2(a)). When S was closed, the fuel-air mixture would be diverted to the tube and the timer would start. Since the flow out of the bottom of the tube caused a pressure drop in the ignition tube, a check-valve (fig. 1) was connected to the top of this tube to keep the pressure inside atmospheric. A heater was also added at the check valve so that the incoming air would not cool the tube excessively. The tube temperatures recorded were those just prior to the run.

The delay was determined as the time between the introduction of the fuel into the mixer (or the closing of the fuel-air-bypass valve) and the appearance of a pressure pulse indicated by the strain-gauge pressure pickup (fig. 1) located at the downstream end of the tube.

Fuel-bypass runs proceeded in the following way: air was allowed to flow through the tube between runs while the fuel flow was bypassed as shown in figure 2(c). The fuel solenoid valve was then energized diverting the fuel into the tube, as in figure 2(b), and the timer started. The fuel valve was shut off after the appearance of the pressure pulse.

For the fuel-air-bypass method, air was allowed to flow through the ignition tube until a few seconds before each run when the lower exhaust and fuel valve were opened. Flow was allowed to proceed until the temperatures of the tube and mixer had steadied at which time the exhaust valve was closed and the mixture diverted through the tube. Delay was measured as before. Four or five runs were made for each point and the average taken as the ignition delay. Later fuel-air-bypass data were taken with the lower exhaust line approximately doubled in diameter (to 19/64" I.D.), since it was felt that perhaps the smaller line was not providing sufficient exhaust to pull all the mixture away from the ignition tube.

As in the previous work, ignition delay was first determined as a function of flow rate in order to determine the flow rate which gave the minimum delay. Then, in most cases, delays measured subsequently were at or near this flow.

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RESULTS

The results for various fuels are presented separately first and then compared. Fuel-bypass and fuel-air-bypass data are also discussed separately and compared in the cases where both methods were used.

Propane

Effect of flow rate. - Using the fuel-bypass method, ignition delay varied with flow rate as shown in figure 3(a). Mean residence time, calculated as the ratio of tube volume (30" length) to volumetric flow rate, is also included on this plot and all subsequent flow-rate plots. (The 30" length was chosen because this was the highest point at which the temperature was measured.) As a comparison, the data of reference 4 at about the same temperature have been included. As can be seen, the shapes of the curves are similar, although the absolute values are not the same. The minimums of the curves occur at higher flows with increasing propane concentration.

Figure 3(b) shows the results using the fuel-air-bypass system with a small lower exhaust line. As is evident, these curves have no minimums in the range investigated; in fact, some of them are concave downward. This latter result was believed due to premature ignition of the mixture. To try to correct this, a larger exhaust line was substituted, and the results are shown in figure 3(c). The curve was straightened but ignition delay was increased considerably. The idea that a minimum probably does exist at a much higher flow rate is supported by figure 3(d) which shows the flow-rate effect at two temperatures. The low-temperature fuel-air-bypass curve does show a minimum. Unfortunately, the system did not give flows greater than approximately 30 liters per minute, and even at this flow rate no leveling off occurred at the higher temperature. The fact that the low-temperature curves cross the residence-time line results from the method used in calculating this residence time. The calculation assumes constant velocity across the tube, which is not the case, since the fuel-air mixture close to the wall remains in the tube a greater time.

Since the minimums, when they occurred, were at flows of about 8 to 12 liters per minute, all delays were measured at a flow rate of 10 liters per minute. However, it must be remembered that with the fuel-air-bypass system these are not the shortest delays observed.

Effect of fuel concentration. - A plot of ignition delay τ against propane concentration using the fuel-bypass system is shown in figure 4(a) for various temperatures. The slopes of the linear portions vary from -0.71 to -0.94 with no apparent trend, a result similar to those of reference 5. At higher propane concentration, delays depend less on the fuel concentration. At the very low temperature and low concentration, the dependence on fuel concentration is greater.

For the fuel-air-bypass system, figure 4(b) shows that the dependence of τ on propane concentration is very similar to that of figure 4(a) and that the absolute values are about the same. However, the similarity in absolute values results from the fact that the flow-rate curves for the systems cross at or near 10 liters per minute. The slopes vary from -0.68 to -0.93 again with no apparent trend. Here, however, the dependence of τ at higher propane concentration does not fall off as rapidly as in the previous case.

A final comparison of fuel concentration data is shown in figure 4(c). Plotted here are fuel-bypass and fuel-air-bypass data, using both a large and small lower exhaust, and data of reference 5. As can be seen, the delays vary at most by a factor of approximately 1.7. The slopes vary from -0.74 to -1.05, values which lie in the range of slopes observed previously. Here again, large and small lower exhaust lines give differing results for the fuel-air-bypass system, but the surprising result obtained is that the size of the lower exhaust also affected the fuel-bypass data. The trends observed, however, remain the same regardless of the procedure used. These can be expressed in an equation of the type

$$1/\tau = \text{Constant } (C_F)^n \quad (1)$$

where C_F is fuel concentration and n is the negative of the slopes of the plots.

Effect of oxygen concentration. - In figure 5 the ignition delay is plotted against oxygen concentration for various concentrations of propane using both the fuel-bypass and fuel-air-bypass methods. The oxygen-nitrogen mixtures used were 9.8, 20.9 (air), 29.5, and 49.9 percent oxygen. In most cases the dependence of $\log \tau$ on \log concentration is linear, but with higher propane and lower oxygen percentages, the dependence on oxygen concentration C_{O_2} is much greater. The data can be represented by an equation of the form

$$\frac{1}{\tau} = \text{Constant } \frac{C_{O_2}}{(1 + kC_{O_2})^2} \quad (2)$$

where k ranges from 1.5 to 4.5 and concentrations are in mole fractions.

n-Butane

All data for n-butane were obtained using the fuel-bypass method.

Effect of fuel concentration. - Figure 6 shows the variation of ignition delay with n-butane concentration at various temperatures. The slopes are generally steeper than those for propane, ranging from -0.97 to -1.7 with larger slopes at lower temperatures. Unfortunately, although a flow-rate effect was found for n-butane, the results are not strictly comparable because all the data of the graph are not at the flow rate giving minimum delay or at the same flow rate. However, the trends are the same and give the same relation as the propane data (eq. (1)).

Effect of oxygen concentration. - Figure 7 shows the effect of oxygen concentration on ignition delay for various n-butane concentrations at two temperatures. The oxygen-nitrogen mixtures used were 9.5, 20.9, and 39.5 percent oxygen. At the lower temperature (445° C), the dependence is large, with the slopes varying from -1.0 to -1.4. As the temperature increases to 612° C, however, the dependence decreases, and the slopes range from -0.25 to -0.65, the absolute value increasing with increasing n-butane concentration. This can be expressed by the equation

$$1/\tau = \text{Constant } (C_{O_2})^m$$

where m depends on fuel concentration and temperature. The high-temperature data can also be represented by equation (2), but it does not apply at the lower temperature.

Isobutane

All data on isobutane were obtained using the fuel-bypass method. No flow-rate data are available for this compound, thus the effect of flow rate on ignition delay cannot be shown.

Effect of fuel concentration. - Figure 8 shows the variation of ignition delay with isobutane concentration. Again delay decreased with increasing fuel concentration. Comparison with figure 6 shows the effect of branched chain structure on delay, which is in general longer for isobutane than for n-butane. Also the slopes of figure 8 vary from -0.73 to -0.93 (increasing with temperature), values which are considerably lower than those for n-butane. The relation between τ and fuel concentration can be represented by equation (1).

Effect of oxygen concentration. - Figure 9 shows ignition delay as a function of oxygen concentration for various isobutane concentrations. The oxygen-nitrogen mixtures contained 9.5, 20.9, and 50.0 percent oxygen. The dependence is much stronger at low oxygen concentrations and increases slightly at higher isobutane concentration. The dependence fits equation (2), where k is approximately 2.

Ethane

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Effect of flow rate. - Using the fuel-bypass method, it was found that for ethane, ignition delay varied with flow rate as it did for propane (fig. 10(a)). Using the fuel-air bypass system, the results again were similar to the same case for propane, and these are shown in figure 10(b) at two temperatures along with data for the fuel-bypass method. The low-temperature fuel-air-bypass curve shows a minimum, while the higher temperature curve does not; again it is believed that a minimum occurs at a much higher flow. Since the minimums (of those curves having them) occurred at 6 to 10 liters per minute, all further data discussed were run at 10 liters per minute.

Effect of fuel concentration. - Figure 11(a) shows the effect of ethane concentration on ignition delay at five temperatures. These data were all obtained using the fuel-bypass method. The effect is one of decreasing delay with increasing concentration. The slopes of the lines vary from -0.90 to -1.25, the dependence of delay on concentration being stronger than that observed for the corresponding propane data. Equation (1) describes the results.

The results using the different procedures are shown in figure 11(b). The size of the lower exhaust not only affects the fuel-air-bypass data but also the fuel-bypass work. However, it is again stressed that the trend, that is, decrease of delay with increasing fuel concentration, did not appear to be affected by the apparatus.

Hydrogen

Effect of flow rate. - Figure 12 shows the effect of flow rate on ignition delays for hydrogen at two temperatures using the small exhaust with both fuel-bypass and fuel-air-bypass methods. The behavior for hydrogen is similar to that observed for propane and ethane in that at the lower temperature both curves show a leveling off. Also, at the higher temperature the fuel-bypass data do level off, but the fuel-air-bypass data do not. The minimums occur at higher flow rates than those for the hydrocarbons.

Effect of fuel concentration. - The effect of fuel concentration on ignition delay is shown in figure 13(a) and again delay decreases with increasing fuel concentration. Both fuel-bypass and fuel-air-bypass data for the small exhaust are plotted. The dependence is stronger at the higher temperature, with the slopes of the lines varying from -0.69 to -1.3. Therefore, equation (1) applies and n is apparently temperature dependent.

Figure 13(b) shows the variation in the delays measured by the different procedures. For hydrogen, the fuel-bypass results with the large and small lower exhaust both fall on about the same line as would be expected (this was not observed with the hydrocarbons). With the fuel-air-bypass data, however, the size of the lower exhaust had a large effect on the delays, increasing them by a factor of 2.

DISCUSSION

Comparison Among Fuels

Figure 14 shows the variation of ignition delay with temperature for all the fuels studied using the fuel-bypass system. Figure 14(a) compares the fuels at a constant concentration (10 percent) which is rich for all fuels except hydrogen. Figure 14(b) compares them at stoichiometric concentration. (The data for this figure were usually extrapolated from the curves of τ against fuel concentration except for hydrogen.) No dependence on oxygen concentration was taken into account, since the data were not available for all compounds.

Figure 14(a) shows that delays at the lower temperatures vary inversely with the oxidizability (the ease with which the fuel combines with oxygen at low temperatures), which increases as the normal paraffin series is ascended and is less for a branched chain than for the corresponding straight-chain compound (ref. 6). The delays also are in proper order with the lowest measured spontaneous ignition temperatures tabulated in reference 7. At the higher temperatures all of the fuels tend to group together. The apparent activation energies which can be obtained from the slopes of the linear portions of the curves are:

| Fuel | Apparent activation energy, kcal |
|-----------|---|
| Ethane | 25 |
| Propane | 19 |
| Isobutane | 21 |
| n-Butane | 15 |
| Hydrogen | 26 |

These values for the hydrocarbons are lower than those obtained from burning-velocity measurements.

In figure 14(b) the order of the delays is changed with propane giving the shortest delays (for the hydrocarbons) at the lower temperatures. Hydrogen, however, falls considerably below the others when compared at the same equivalence ratio.

The apparent activation energies calculated from these curves are:

| Fuel | Apparent activation energy, kcal |
|-----------|---|
| Ethane | 27 |
| Propane | 16 |
| Isobutane | 14 |
| n-Butane | 18 |
| Hydrogen | 40 |

The values are low for the hydrocarbons, and the hydrogen value is high.

Effect of Experimental Procedure on Absolute Values of Ignition Delay

In the introduction mention is made of the possibility of fuel cracking and its effect on the results. This effect was first considered when it was noticed that at the higher temperatures the delays measured for the hydrocarbons grouped together. It was thought that perhaps all the hydrocarbons were cracking to about the same products and the differences which might be present were being obscured. In the fuel-bypass system there was a section of the fuel line (fig. 2(c)) between the bypass point and the mixer which contained stagnant fuel at temperatures greater than 1000° F. Rough calculations showed that the fuel cracking could be serious in the time (approximately 1 minute) which was allowed between runs, so the fuel-air-bypass system was tried. Since the fuel-air mixture would be fresh each time, it was hoped that these results would be more meaningful.

Later, as a check on the system, hydrogen was run, and this fuel should have given the same results using either system. Unfortunately, it did not, as is evident from figure 13. For this reason, no conclusions could be drawn about the effect of fuel cracking on the delays. Why the hydrogen delays are not the same using the different procedures is not known, but speculation has led to the belief that perhaps the flow pattern affected the results. Although the tube had screens in the diffuser section to take out the swirl in the flow, these screens burned out rather rapidly. They were only replaced when the system had a major repair, and since it was not known when they were gone, the type of flow present in the ignition tube was unknown. Unfortunately the present system would be difficult to modify in order to determine the type of flow or the flow pattern.

The difference in the type of flow-rate data obtained for the two systems is also hard to explain. It is unfortunate that this system could not go to higher flows where a minimum might have been detected.

Although the experimental technique affected results, the same type of basic relation among ignition delay, concentration, and temperature holds regardless of the particular method used. Delays are inversely proportional to the fuel concentration and the temperature, and are affected by oxygen concentration to some degree depending on the fuel. As would be expected, they depend more strongly on the oxygen concentration when this is low. Although these absolute values may only be true for this particular system, the trends observed are believed to be real.

The results presented here seem to indicate that more research is necessary in this field. The comparison in figure 17 of reference 8 of the ignition delays for hydrogen from several sources shows wide discrepancies which need clearing up. The reader is referred to the discussion on this point in reference 8.

SUMMARY OF RESULTS

An investigation of ignition delays for hydrocarbon and hydrogen mixtures with nitrogen and oxygen showed that absolute values of ignition delay depended on the experimental procedure used but that the trends were the same by either method. Increasing fuel concentration, oxygen concentration, and temperature all decreased the delay as follows:

1. Delays were inversely proportional to the fuel concentration to a power which depends on the fuel and sometimes on the temperature. These exponents ranged in value from 0.6 to 1.7.
2. Delays decreased to a lesser extent with increasing oxygen concentration, for propane and the butanes, the dependence being greater at lower oxygen, higher fuel concentrations.

Lewis Flight Propulsion Laboratory
National Advisory Committee for Aeronautics
Cleveland, Ohio, September 4, 1956

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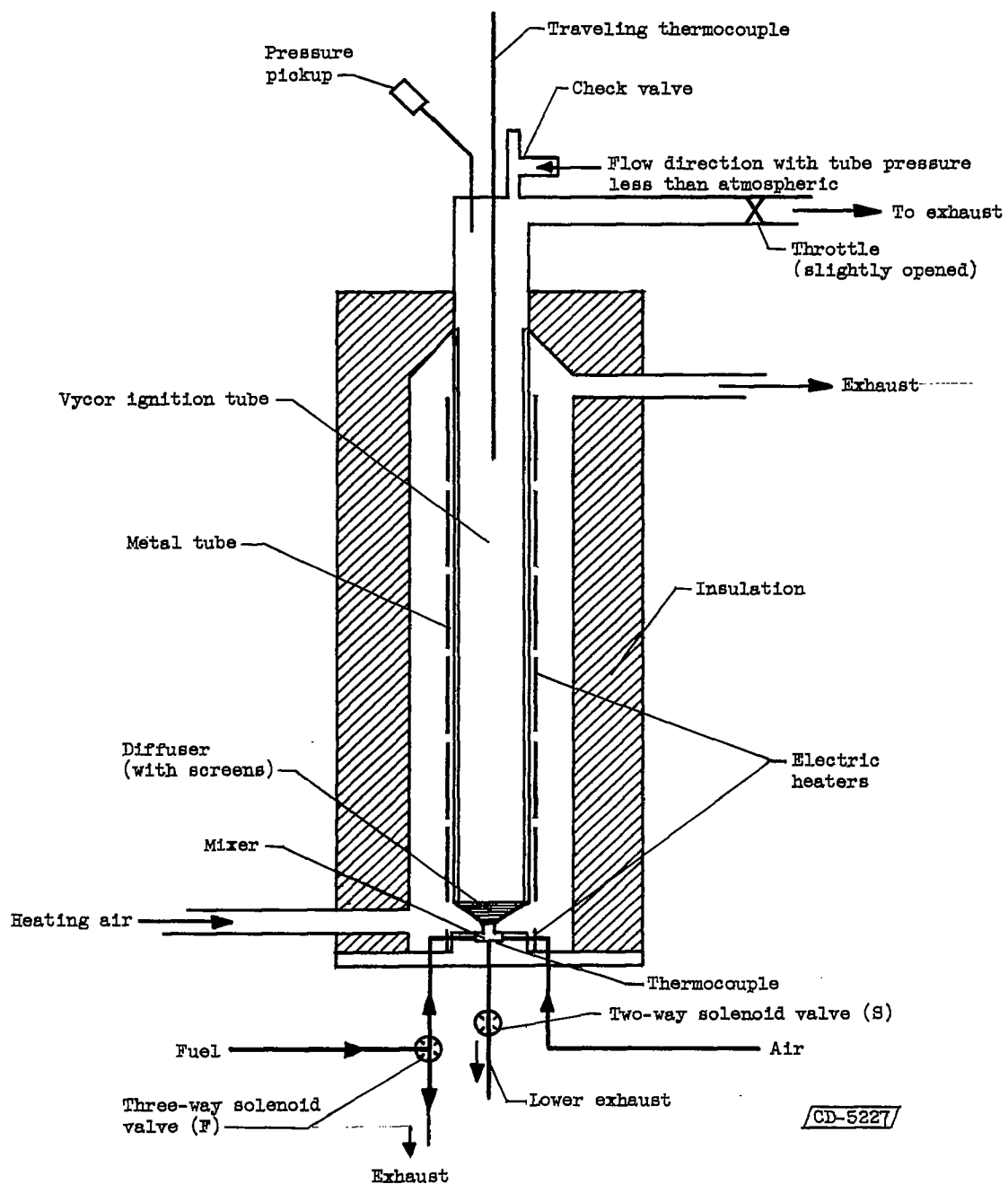


Figure 1. - Schematic diagram of spontaneous ignition delay apparatus.

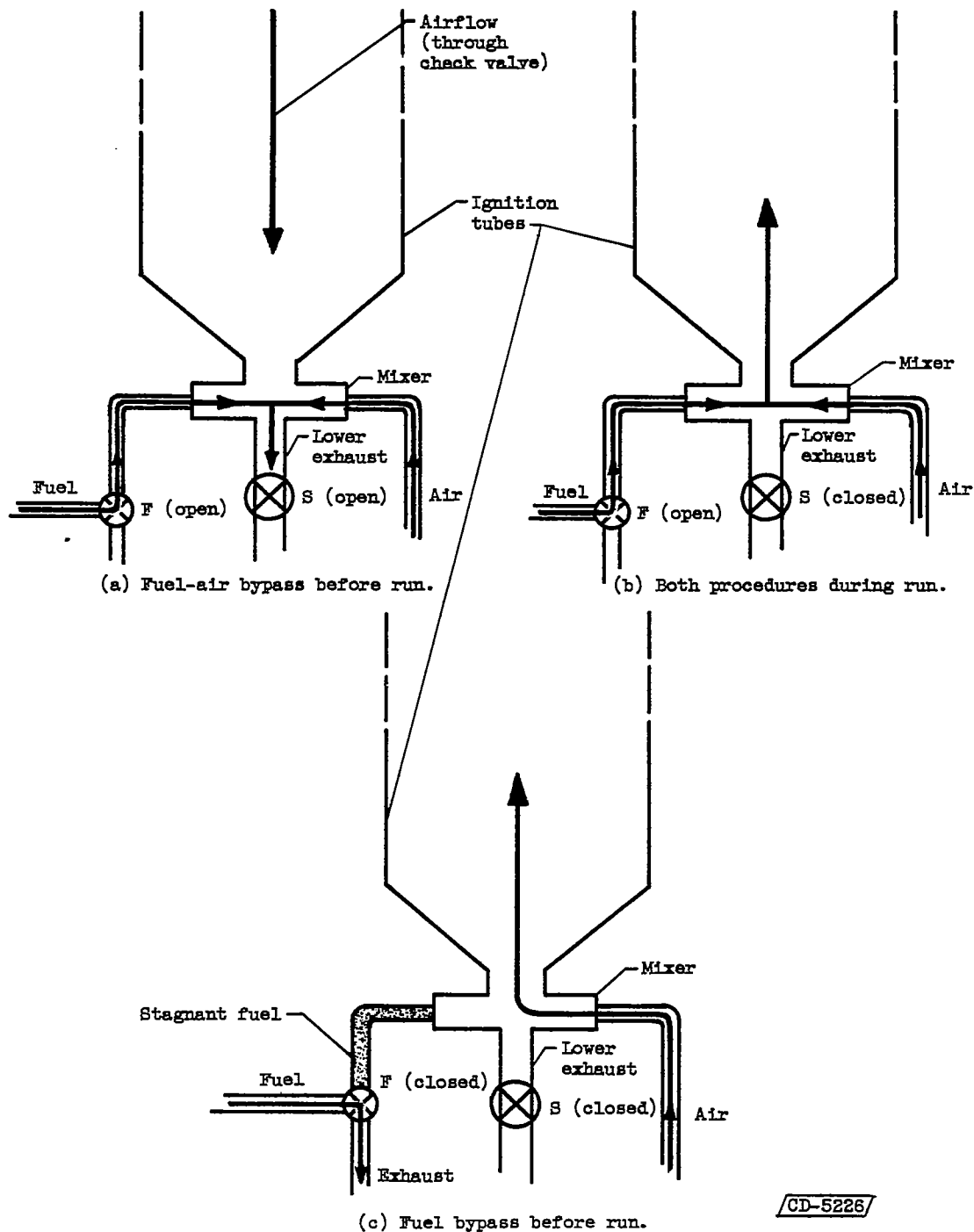
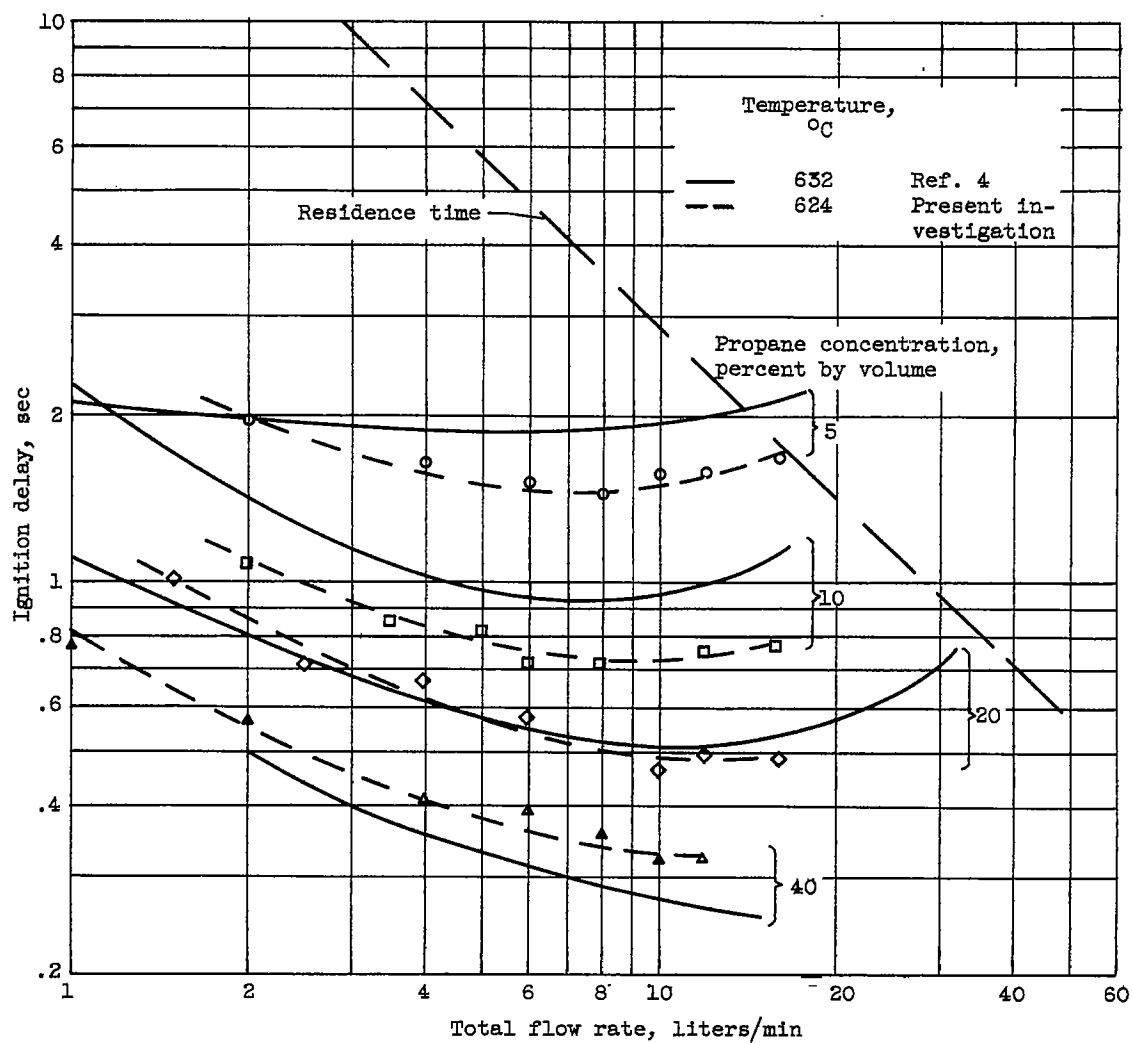
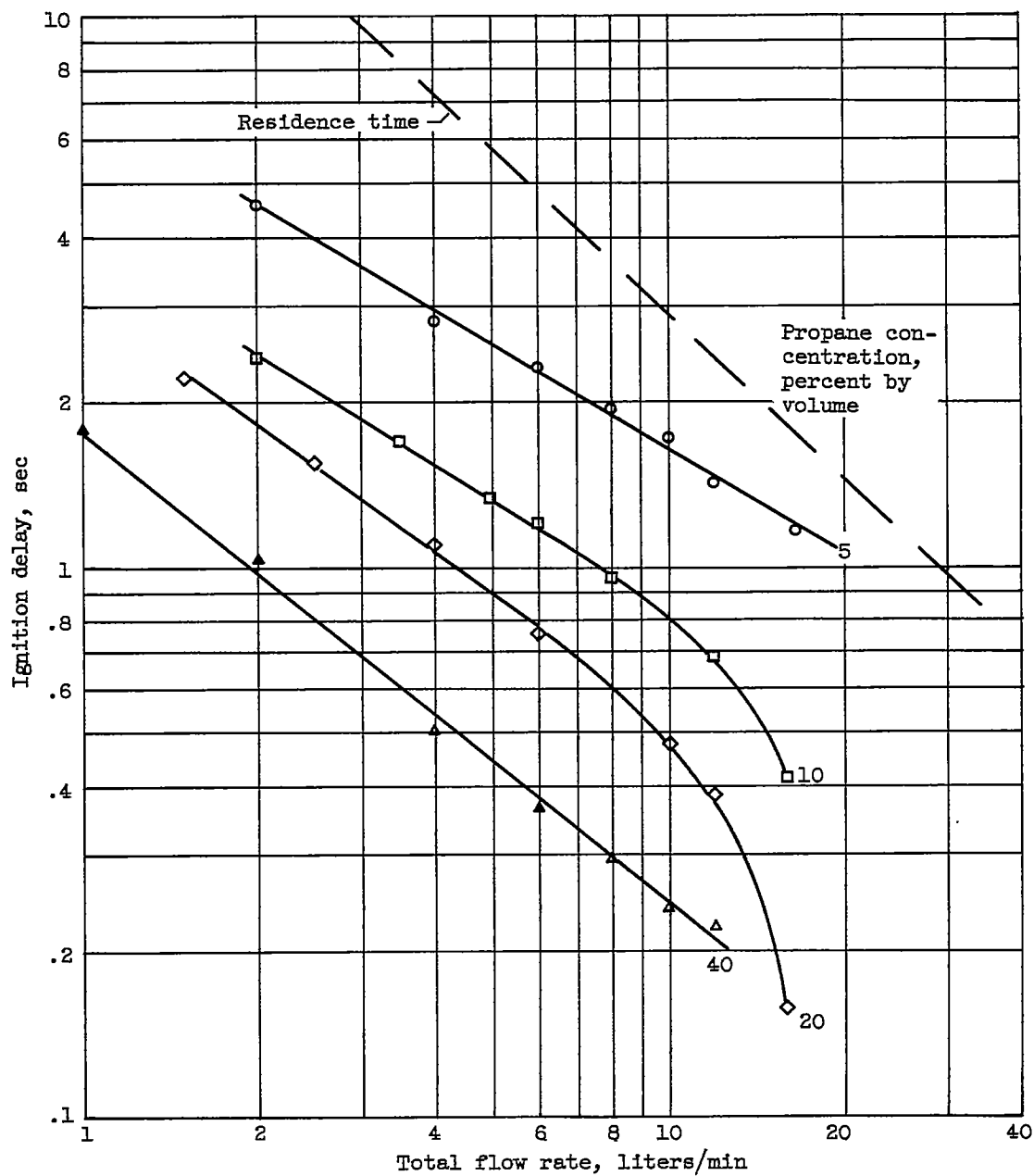


Figure 2. - Diagram of flow systems before and during runs using two procedures.
F, three-way fuel solenoid valve; S, two-way lower exhaust solenoid valve.



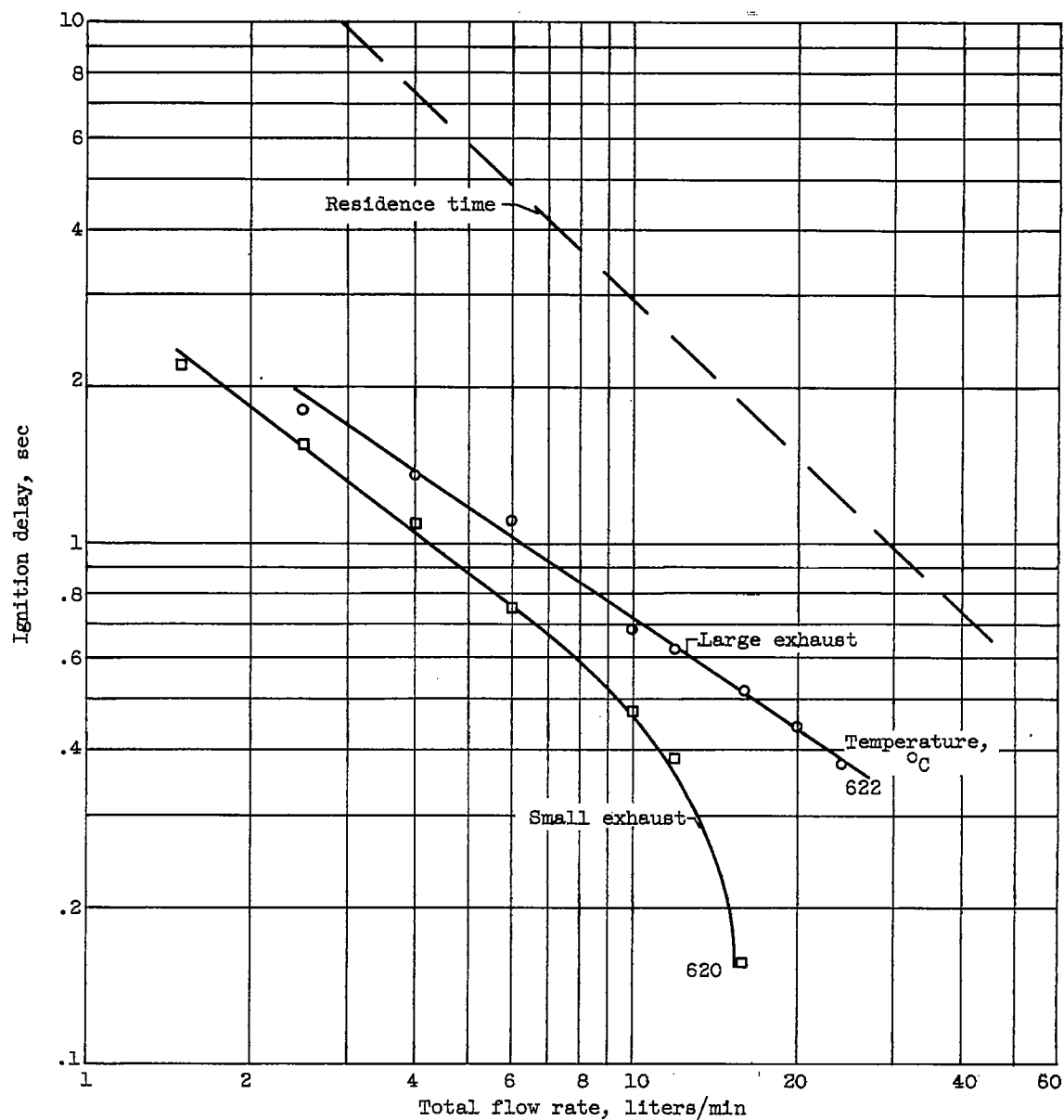
(a) Using fuel bypass.

Figure 3. - Variation of ignition delay with flow rate for propane.



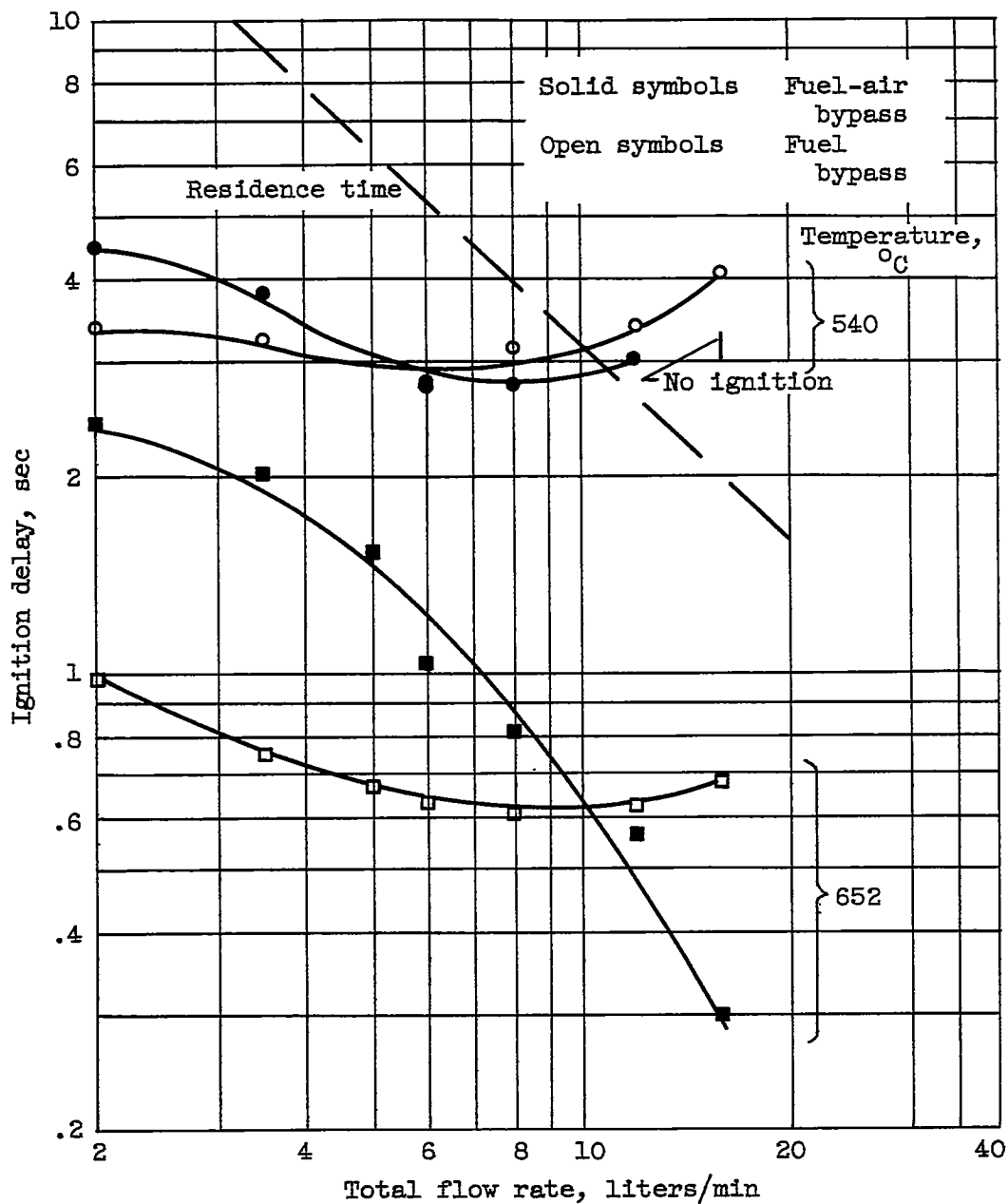
(b) Using fuel-air bypass with small exhaust. Temperature, 623° C.

Figure 3. - Continued. Variation of ignition delay with flow rate for propane.



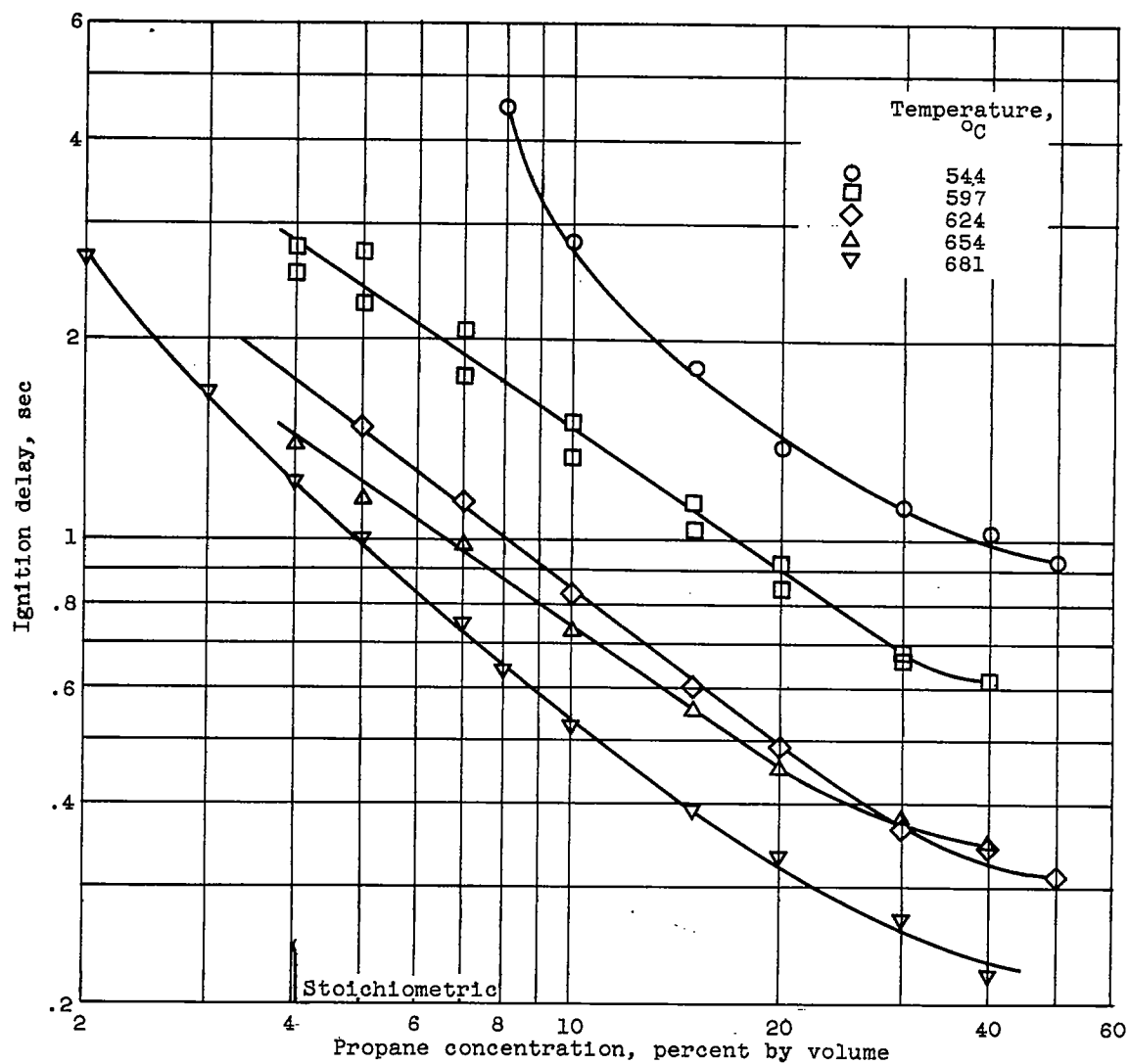
(c) Using fuel-air bypass with large and small exhaust lines. Propane concentration, 20 percent by volume.

Figure 3. - Continued. Variation of ignition delay with flow rate for propane.



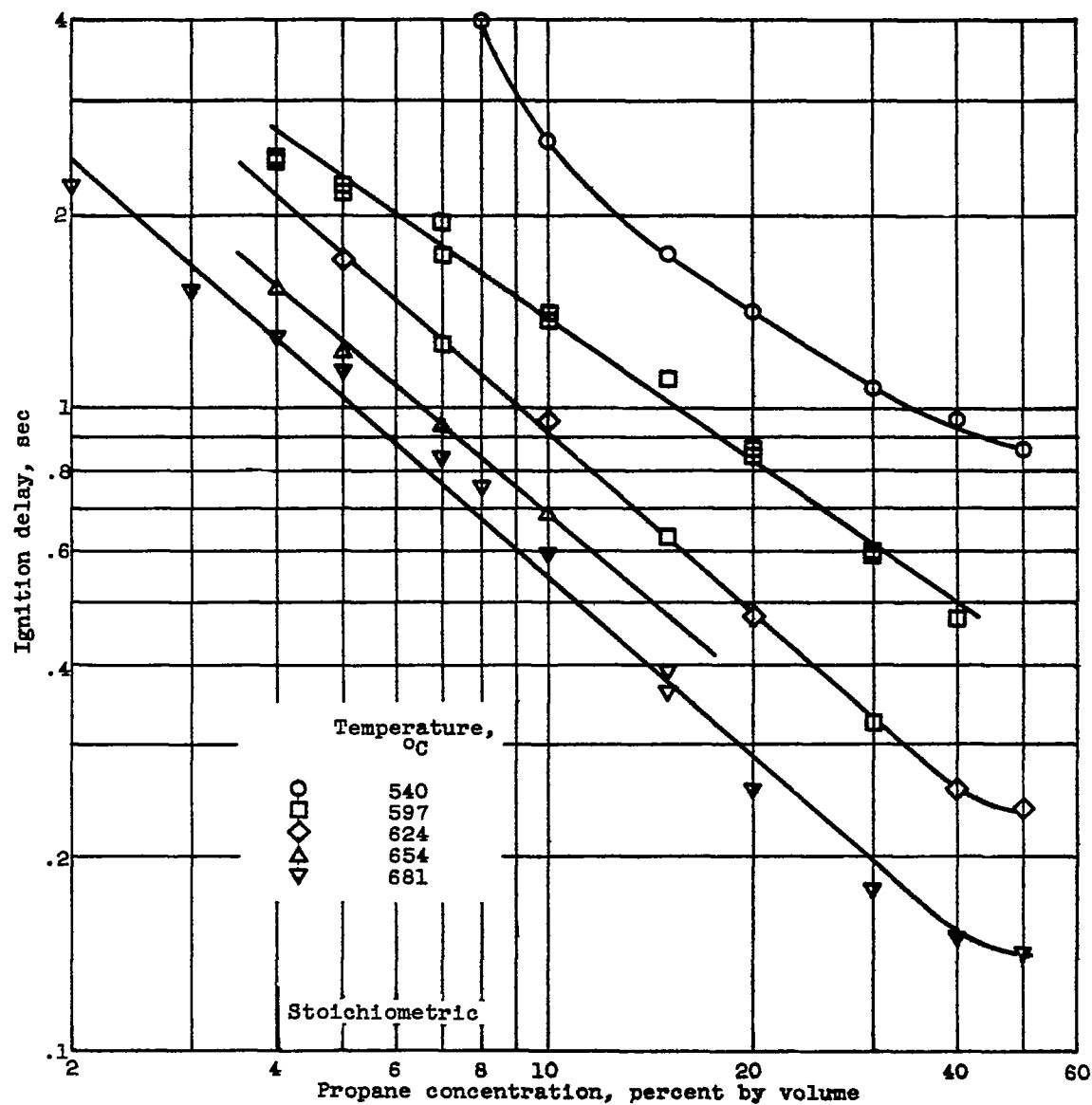
(d) Using fuel-air bypass and fuel bypass. Propane concentration, 10 percent by volume.

Figure 3. - Concluded. Variation of ignition delay with flow rate for propane.



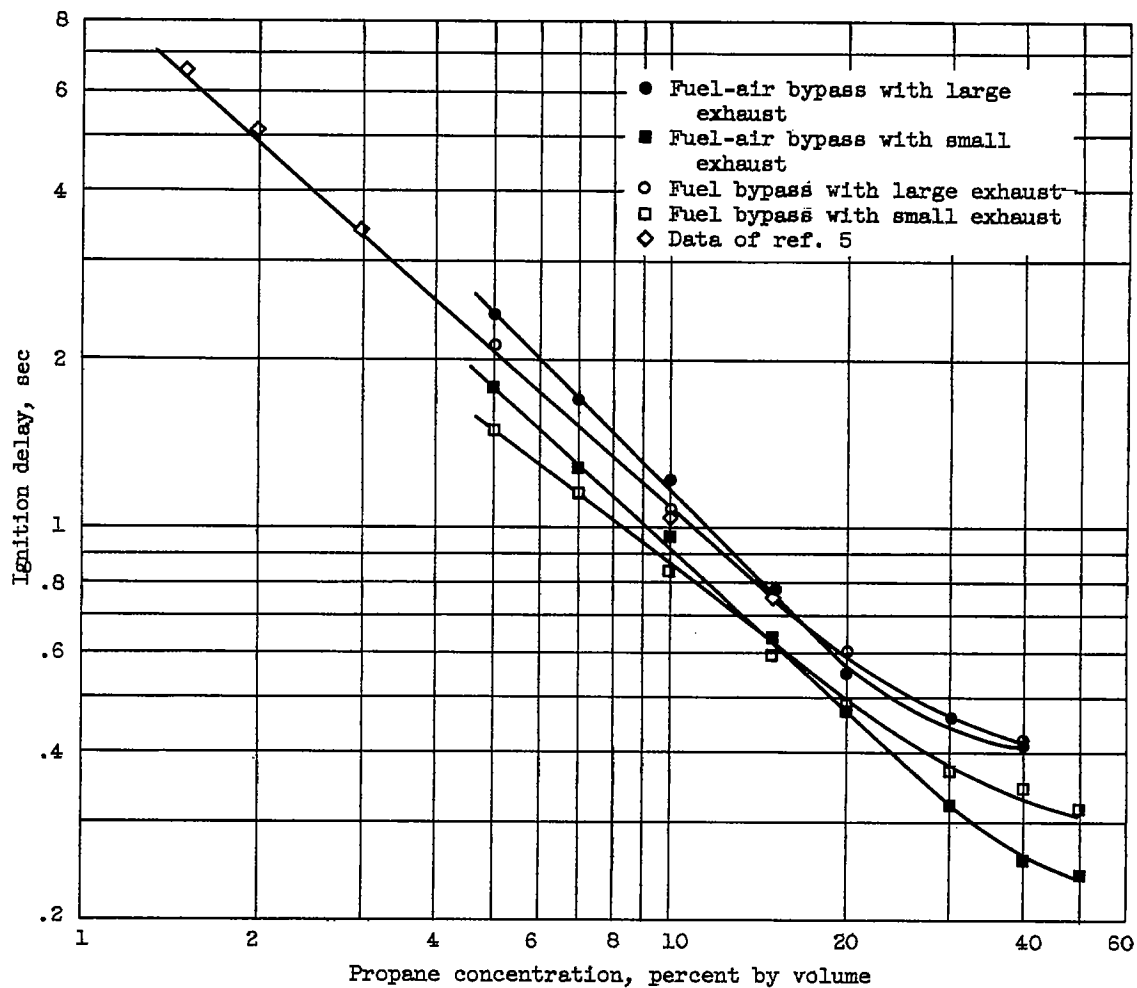
(a) Using fuel bypass.

Figure 4. - Variation of ignition delay with propane concentration.



(b) Using fuel-air bypass with small exhaust.

Figure 4. - Continued. Variation of ignition delay with propane concentration.



(c) Using fuel-air bypass and fuel bypass with large and small exhaust lines.
Temperature, 623° C.

Figure 4. - Concluded. Variation of ignition delay with propane concentration.

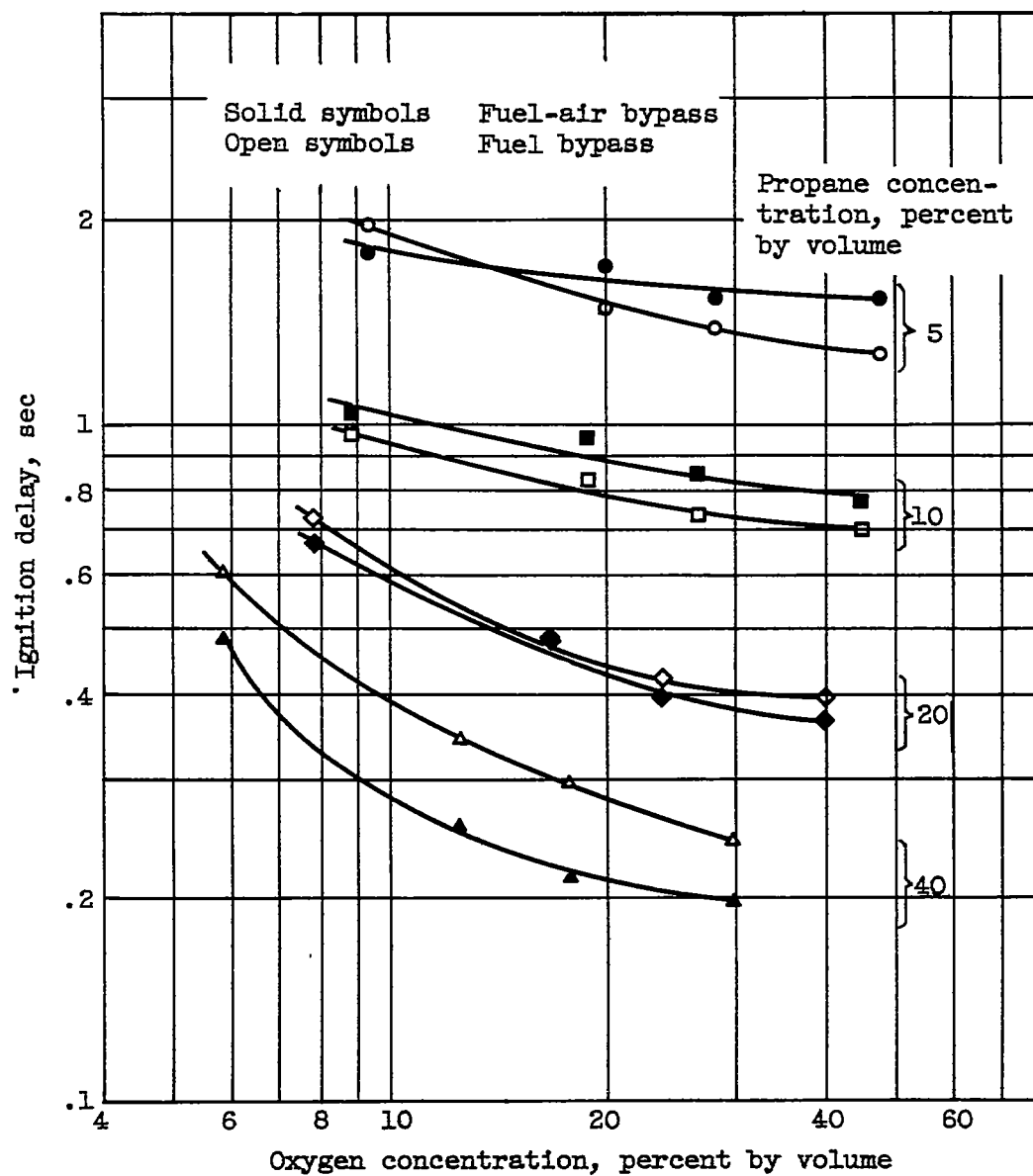


Figure 5. - Variation of ignition delay with oxygen concentration for propane. Temperature, 624° C.

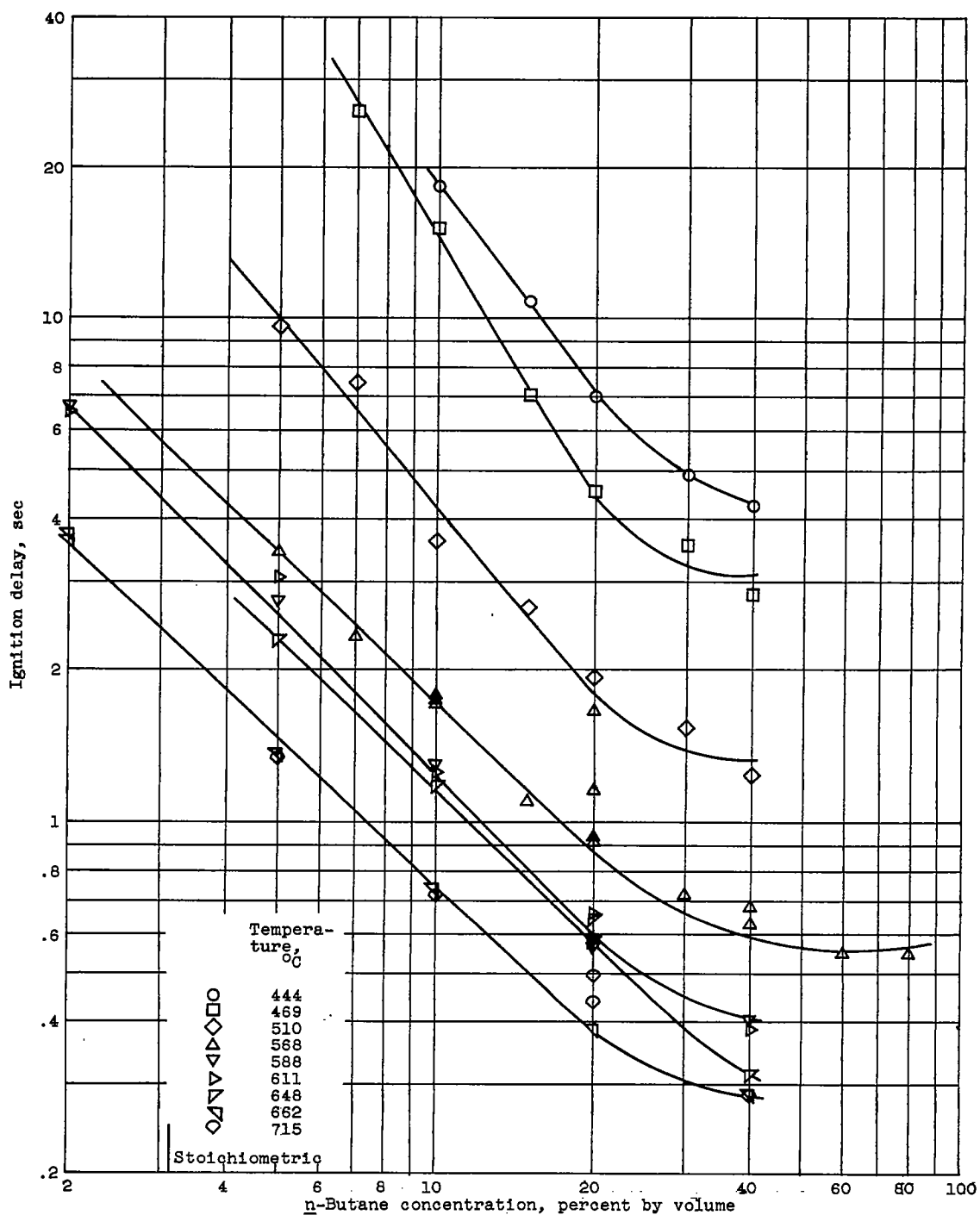
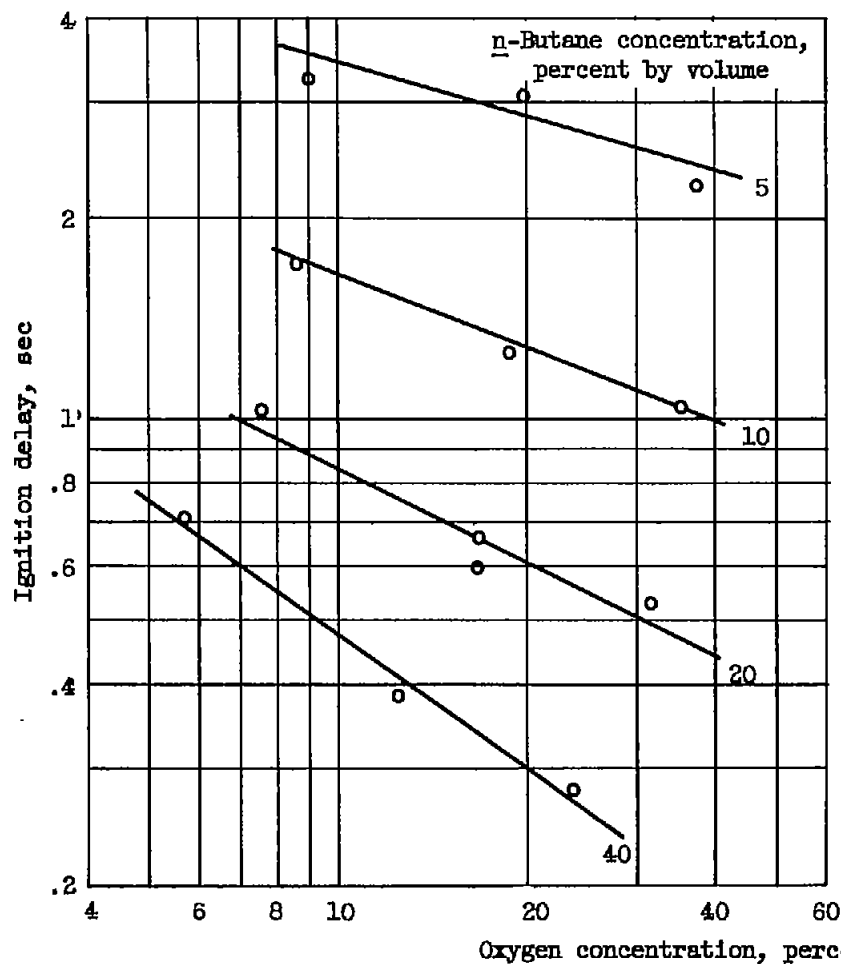
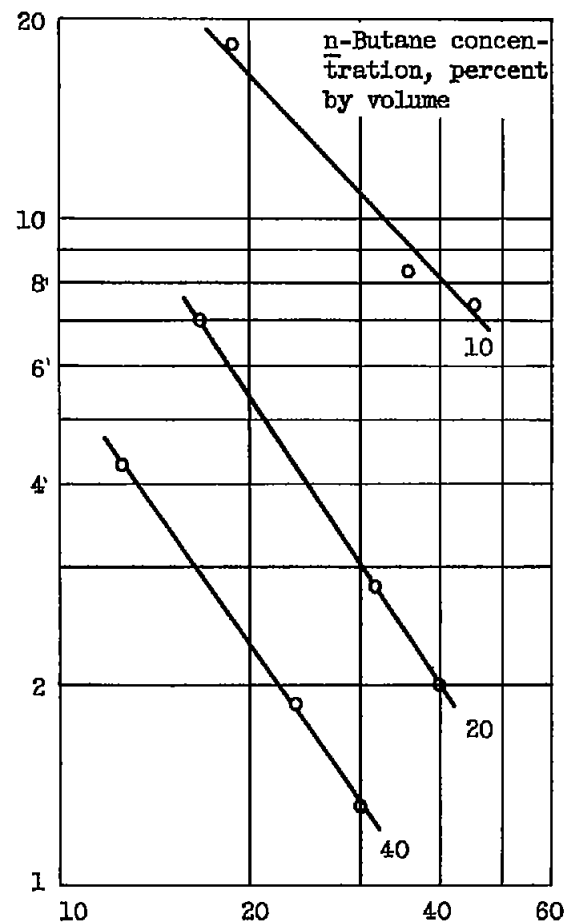


Figure 6. - Variation of ignition delay with n-butane concentration at various temperatures. Fuel-bypass method.



(a) Temperature, 612° C.



(b) Temperature, 445° C.

Figure 7. - Comparison of dependence of ignition delay on oxygen concentration for n-butane at two temperatures. Fuel-bypass method.

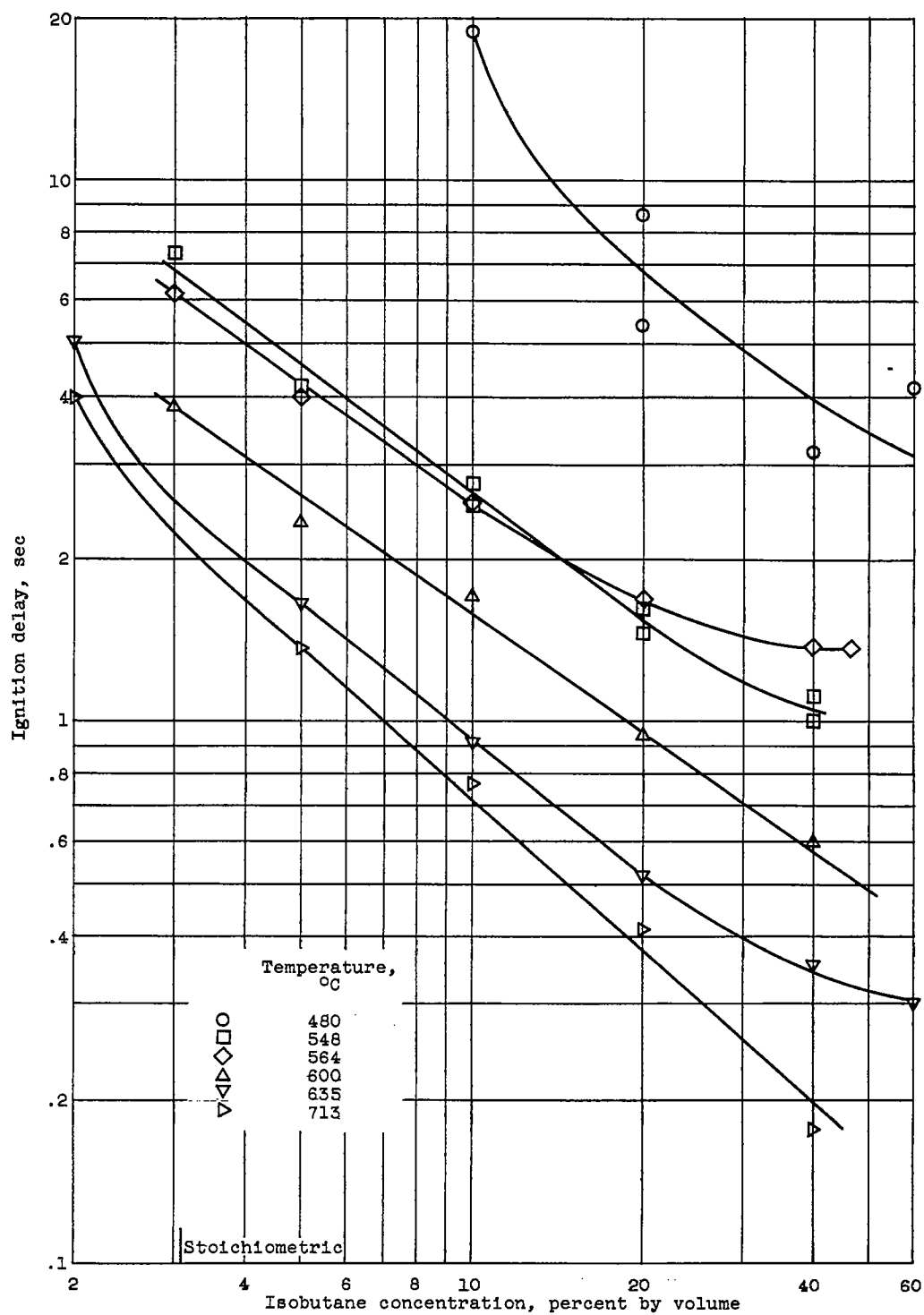


Figure 8. - Variation of ignition delay with isobutane concentration at various temperatures. Fuel-bypass method.

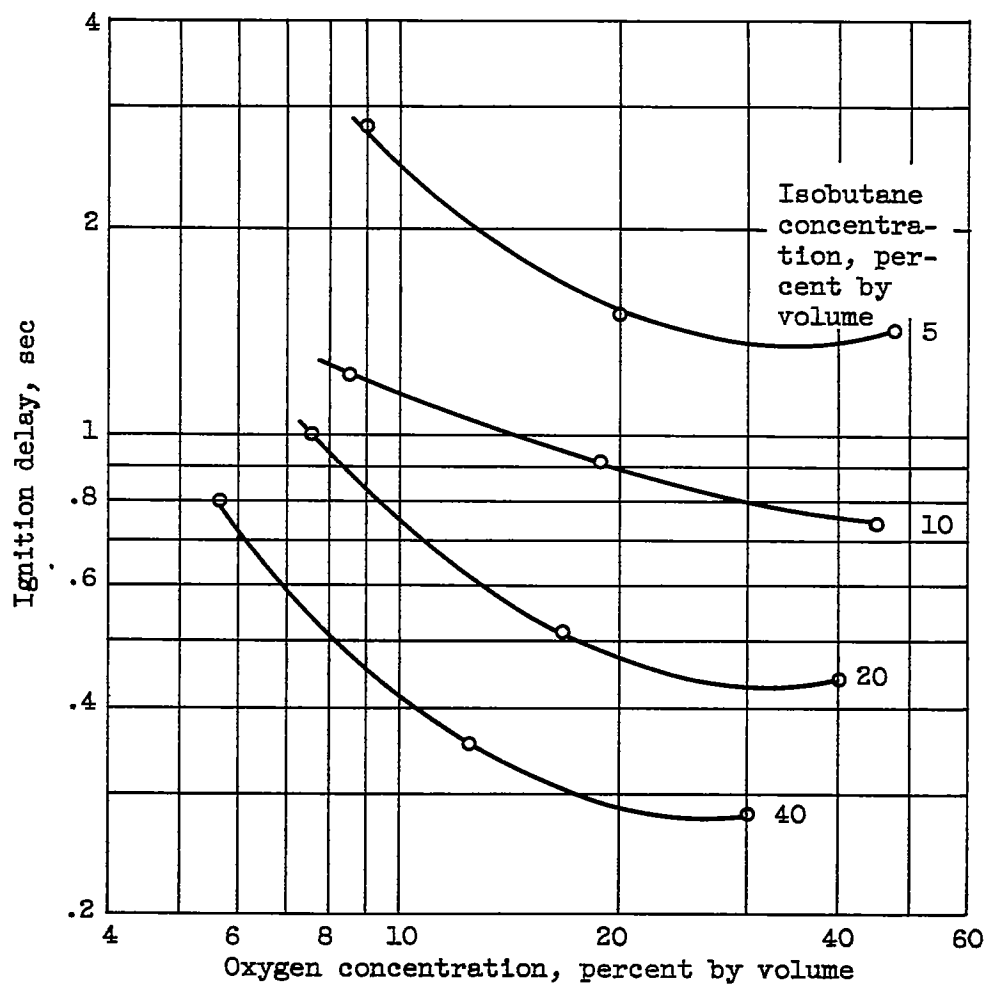
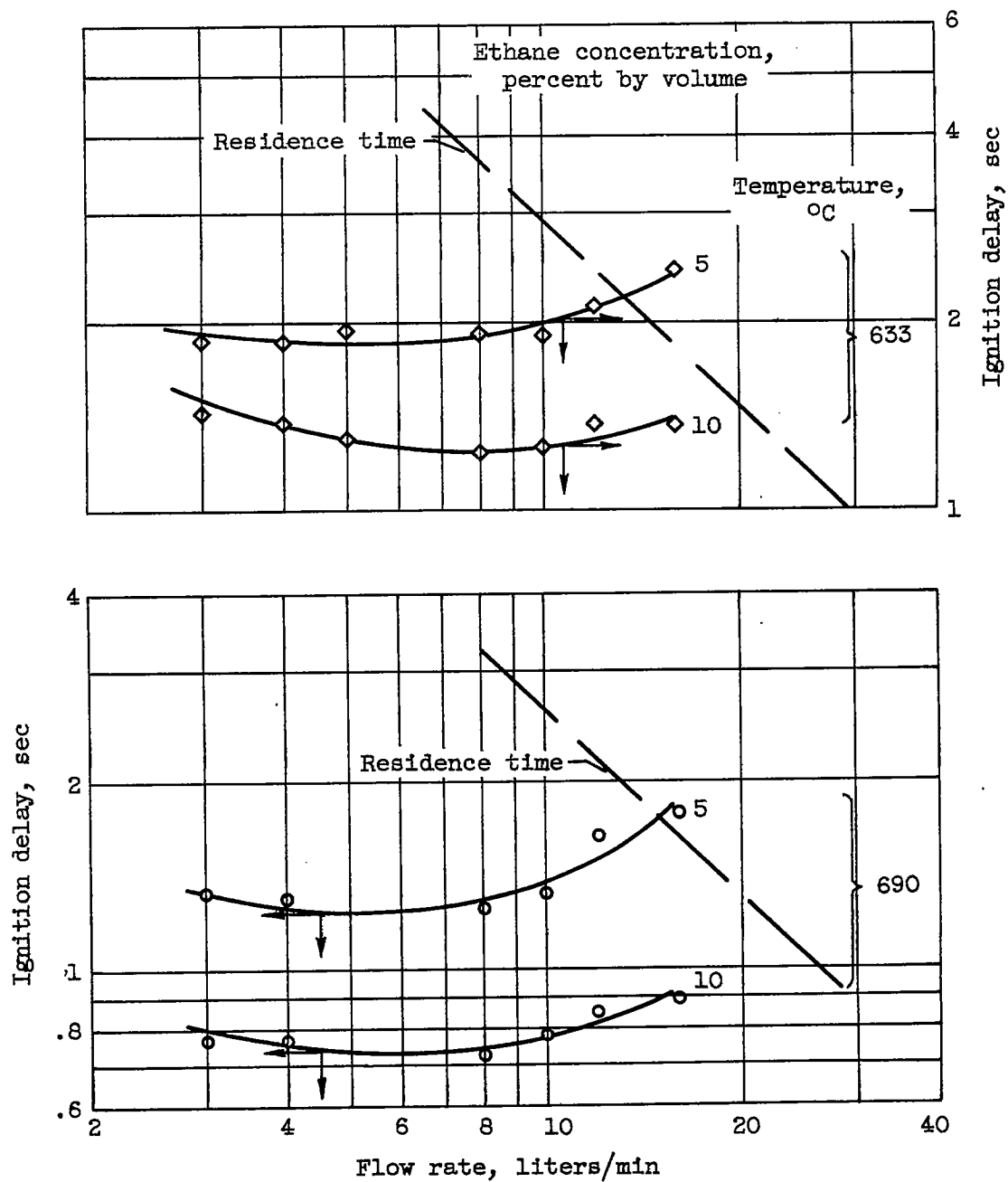
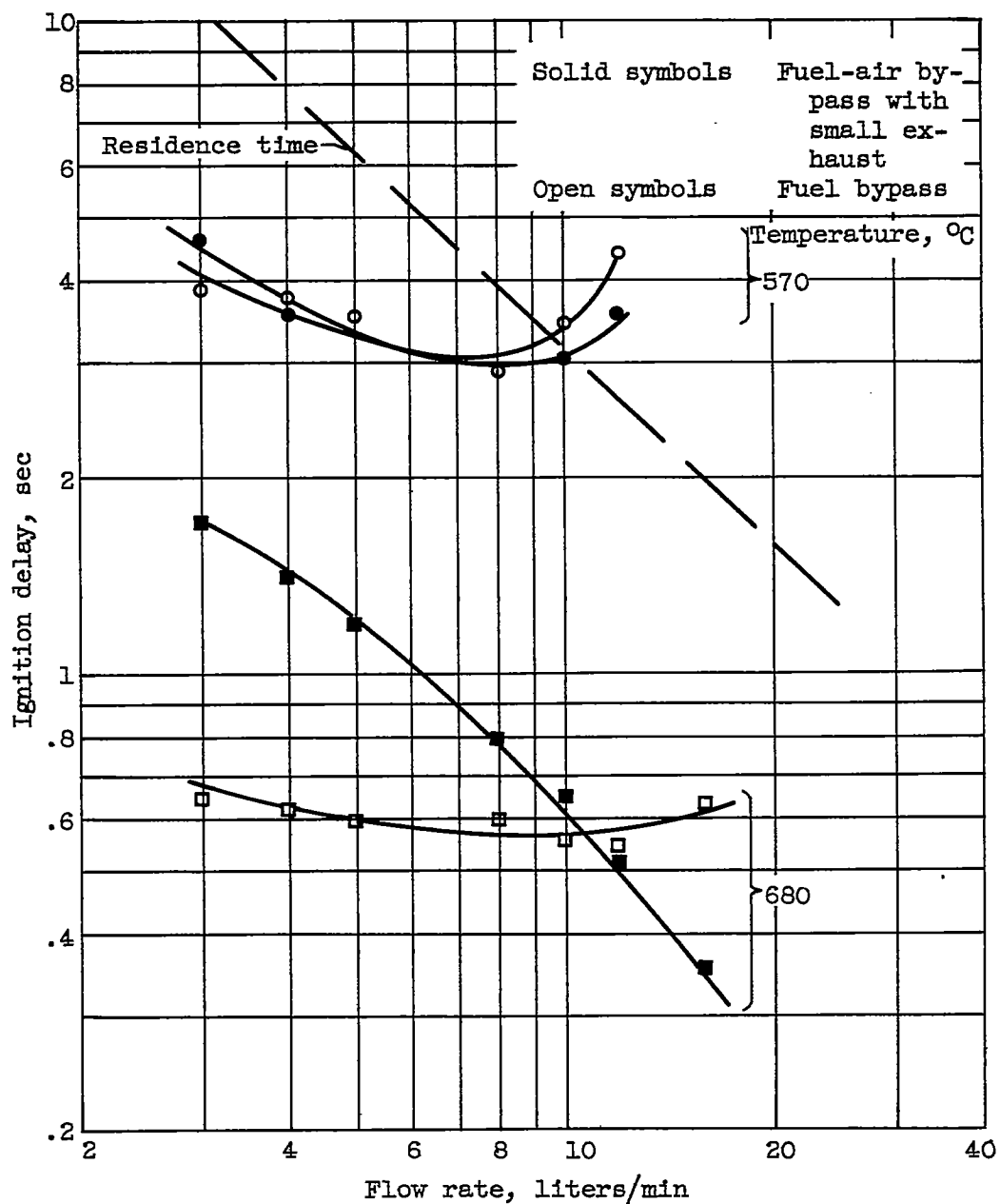


Figure 9. - Variation of ignition delay with oxygen concentration for isobutane.



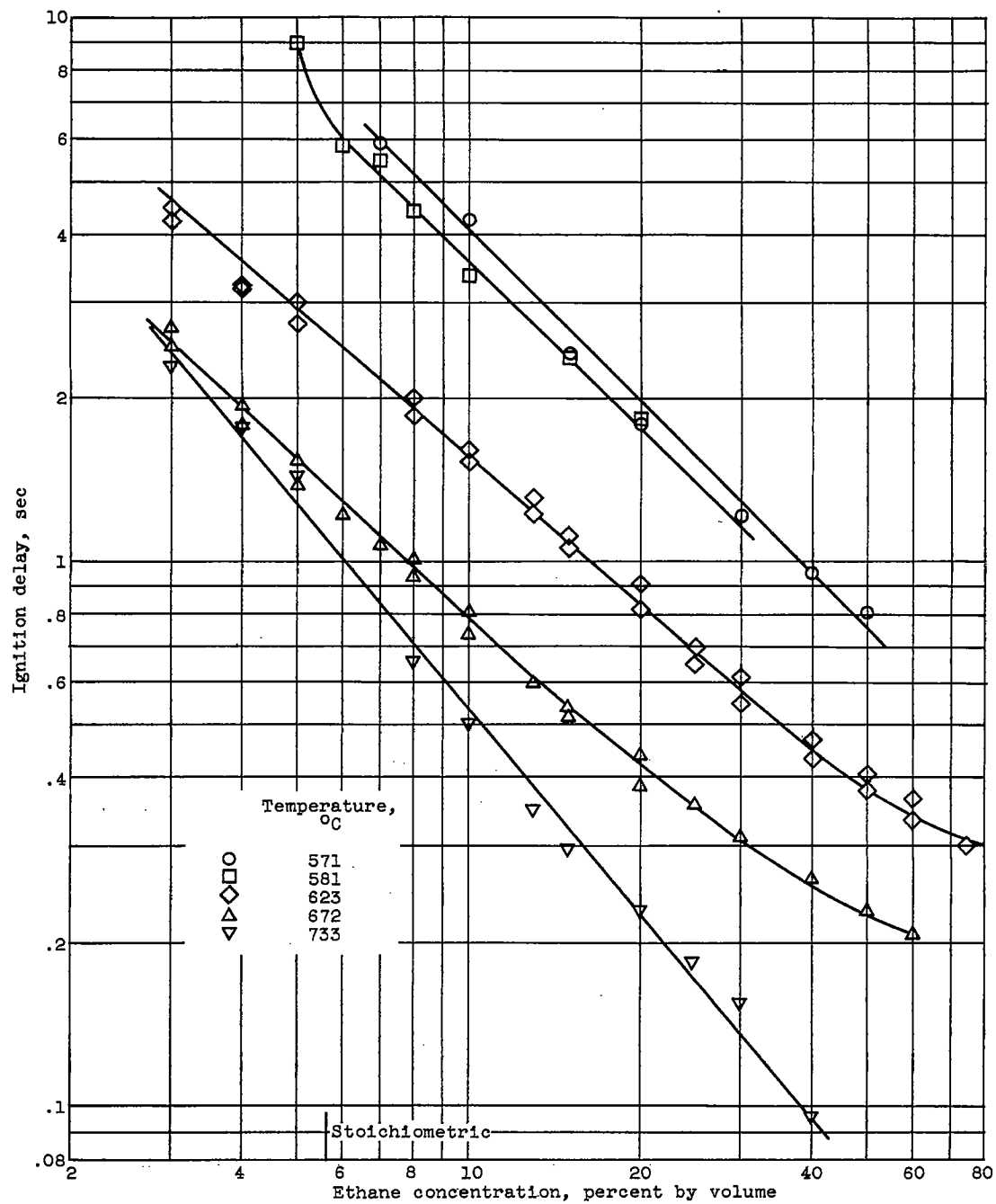
(a) Using fuel bypass.

Figure 10. - Variation of ignition delay with flow rate for ethane.



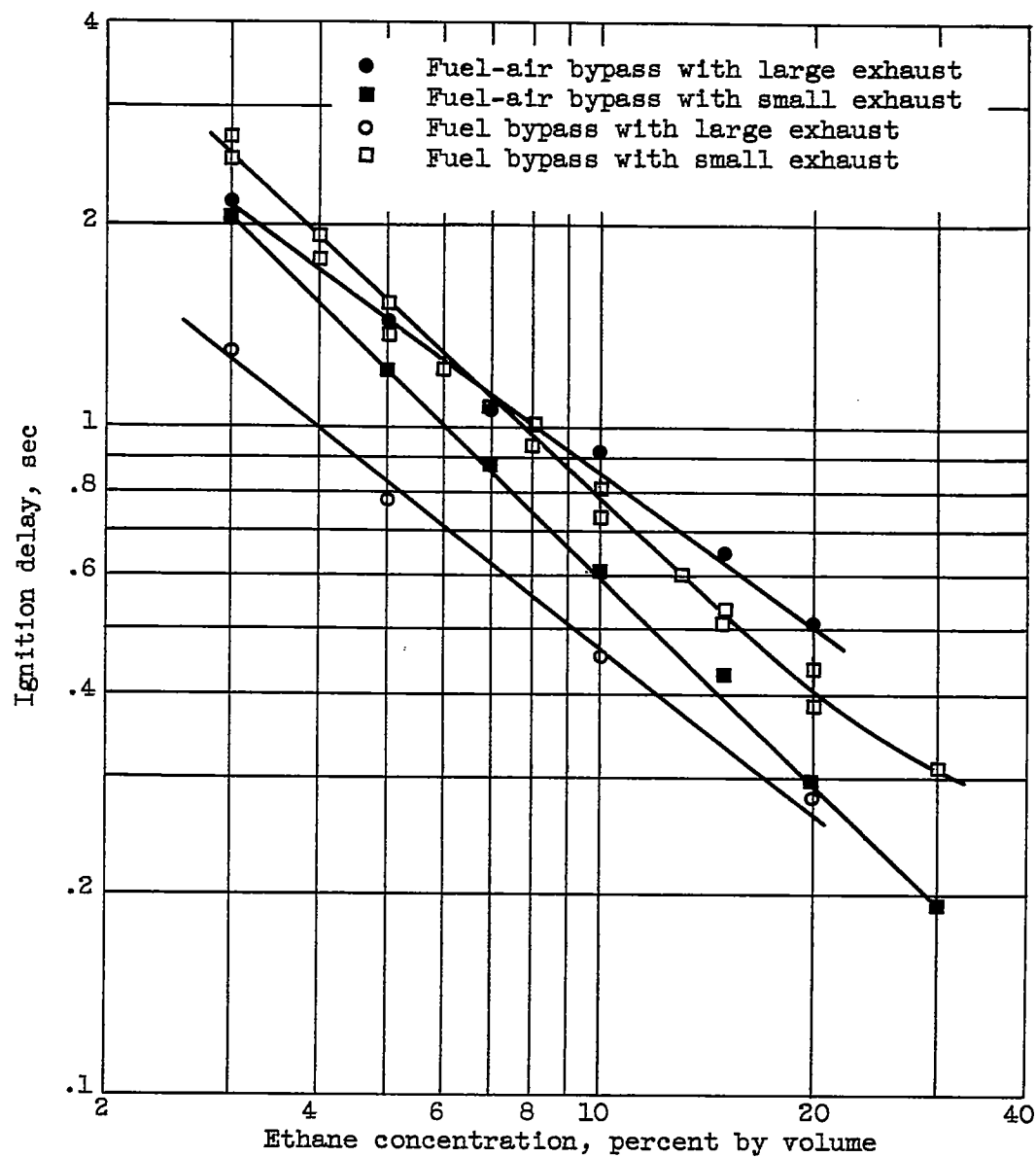
(b) Using fuel-air bypass with small exhaust and fuel bypass. Ethane concentration, 10 percent by volume.

Figure 10. - Concluded. Variation of ignition delay with flow rate for ethane.



(a) Using fuel bypass.

Figure 11. - Variation of ignition delay with ethane concentration.



(b) Using fuel-air bypass and fuel bypass with large and small exhaust lines. Temperature, 678°C .

Figure 11. - Concluded. Variation of ignition delay with ethane concentration.

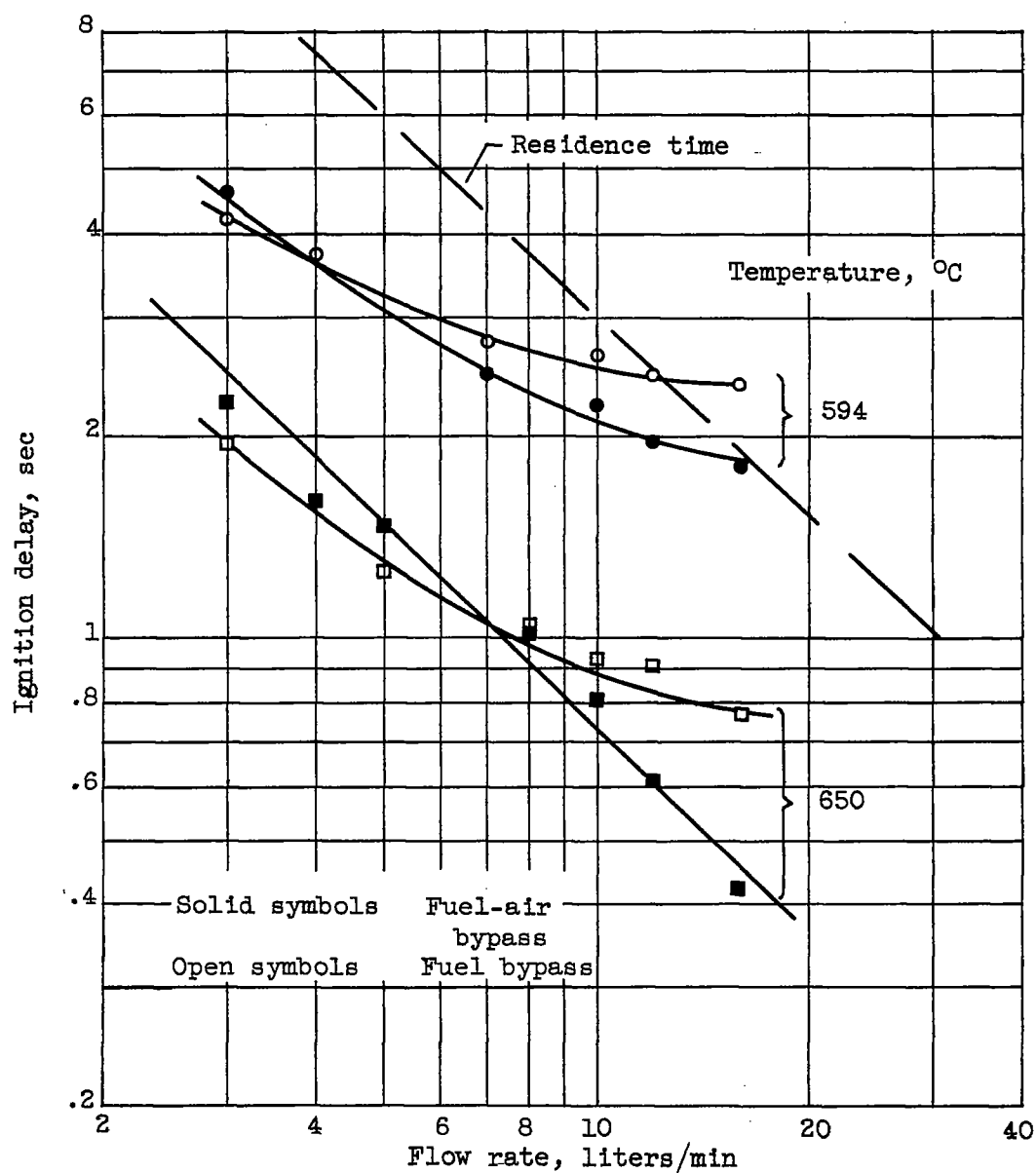
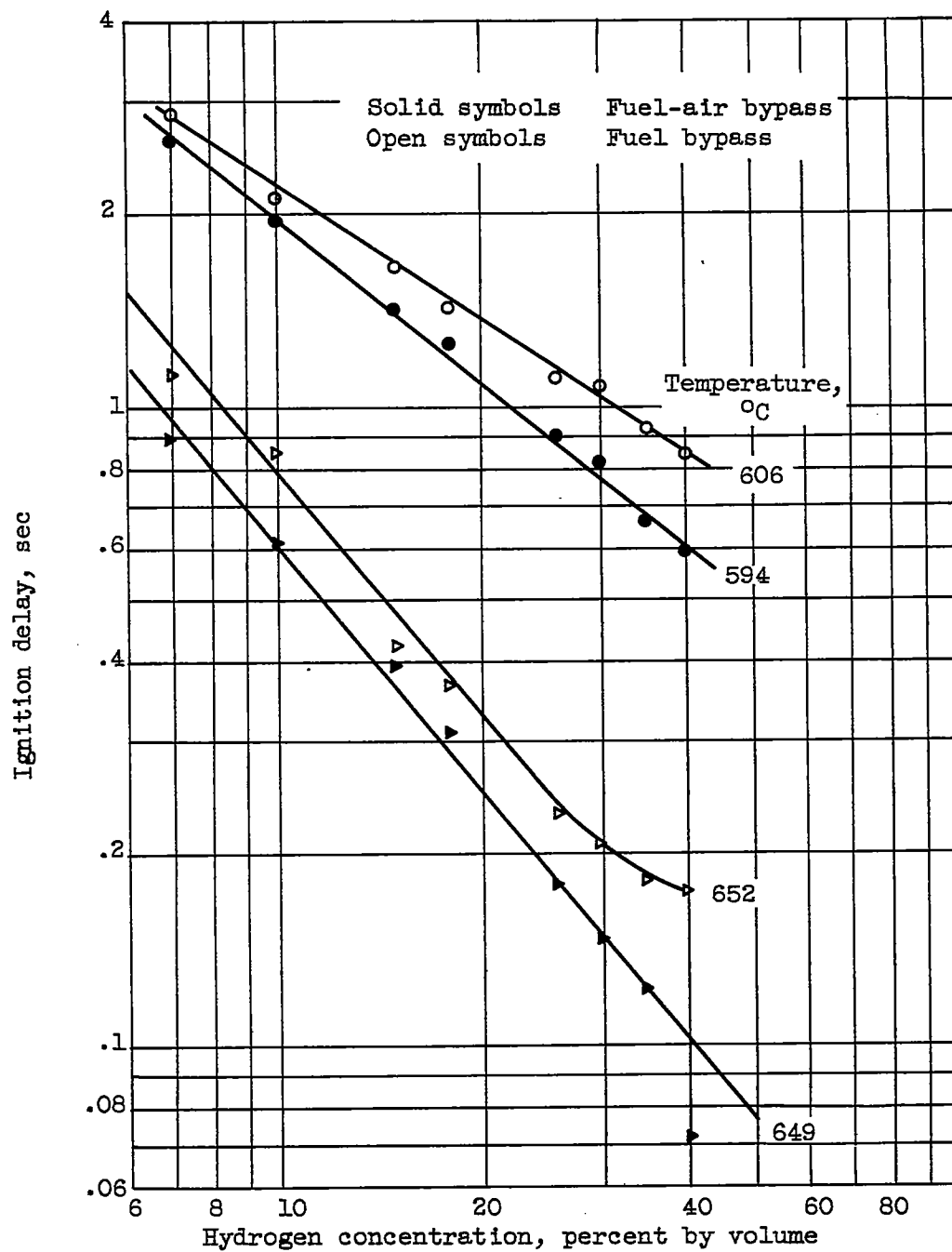
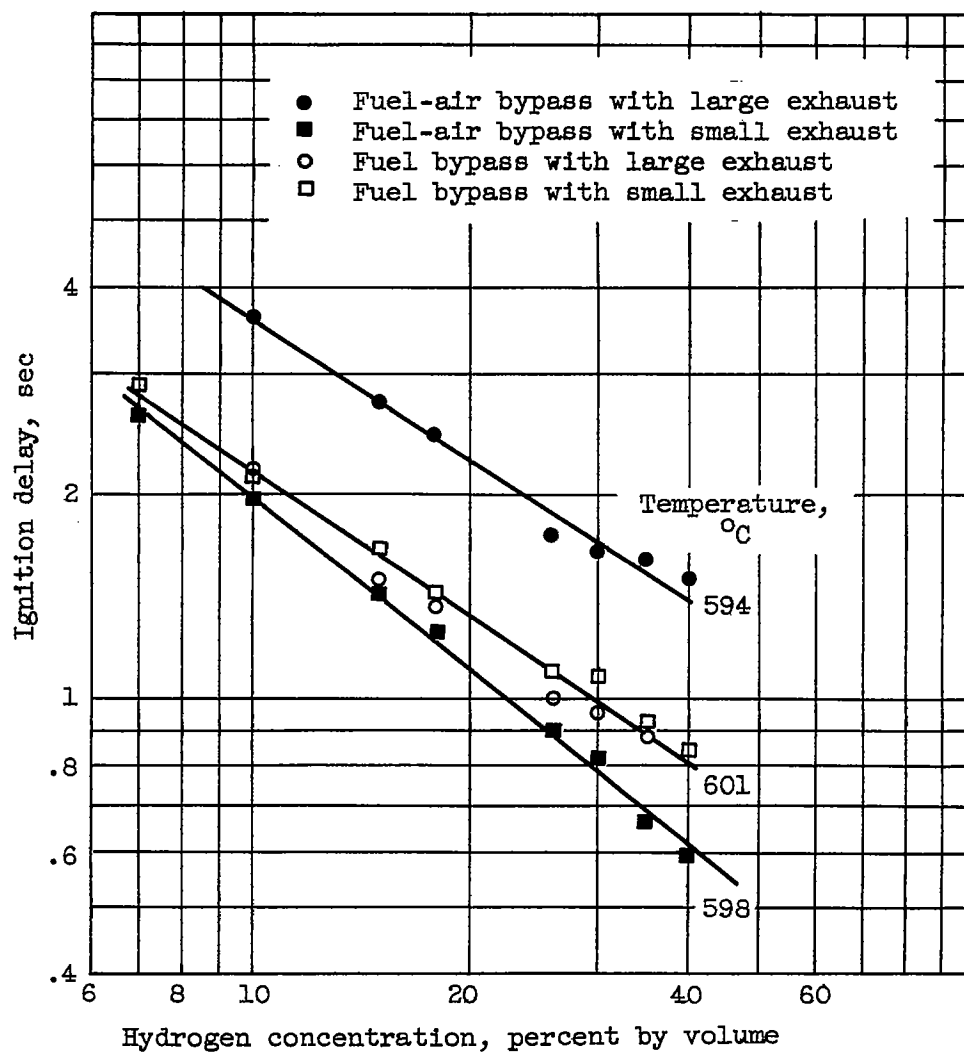


Figure 12. - Variation of ignition delay with flow rate for hydrogen at two temperatures. Hydrogen concentration, 10 percent by volume; small exhaust.



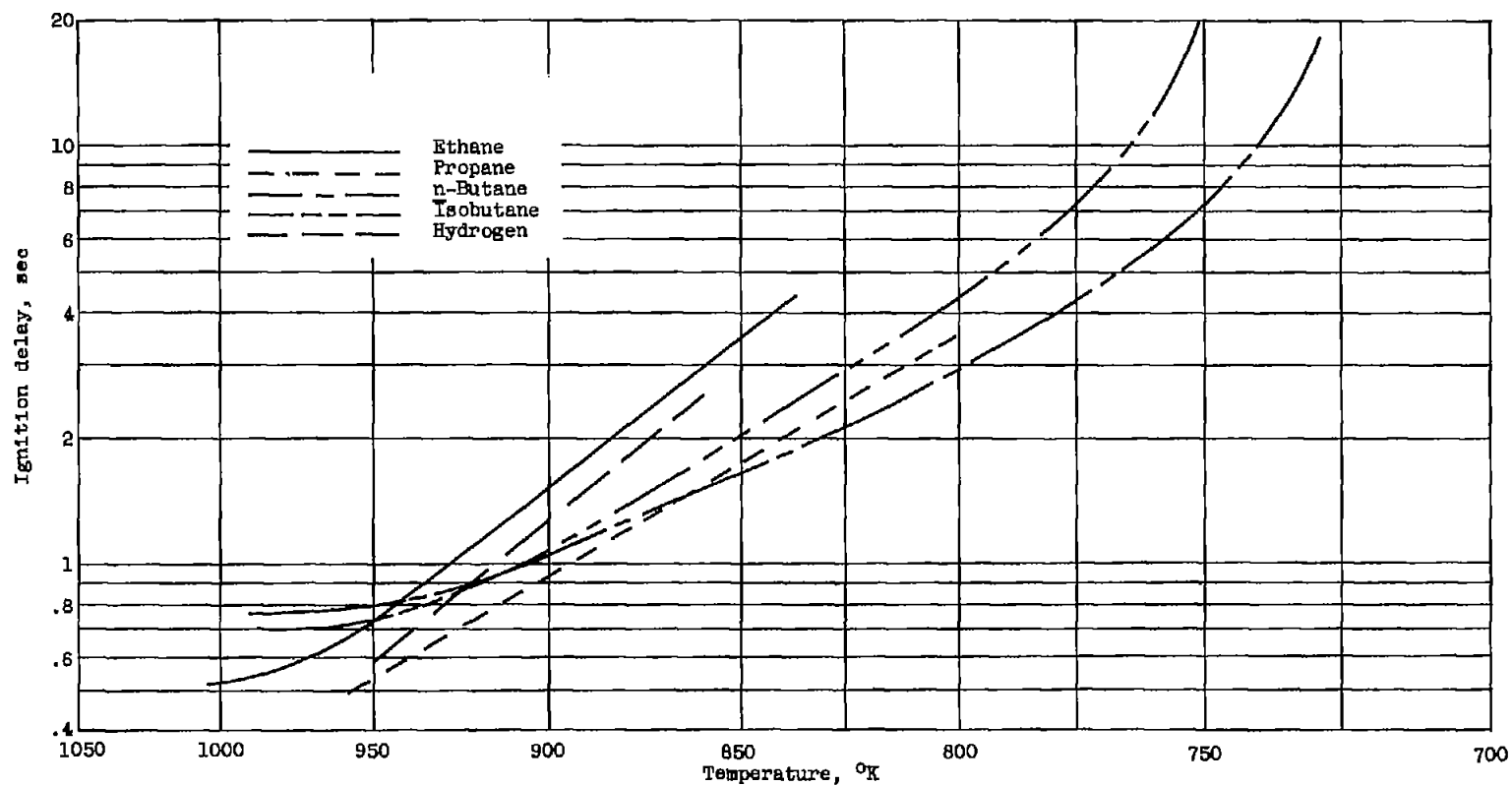
(a) Using fuel-air bypass and fuel bypass with small exhaust.

Figure 13. - Variation of ignition delay with hydrogen concentration.



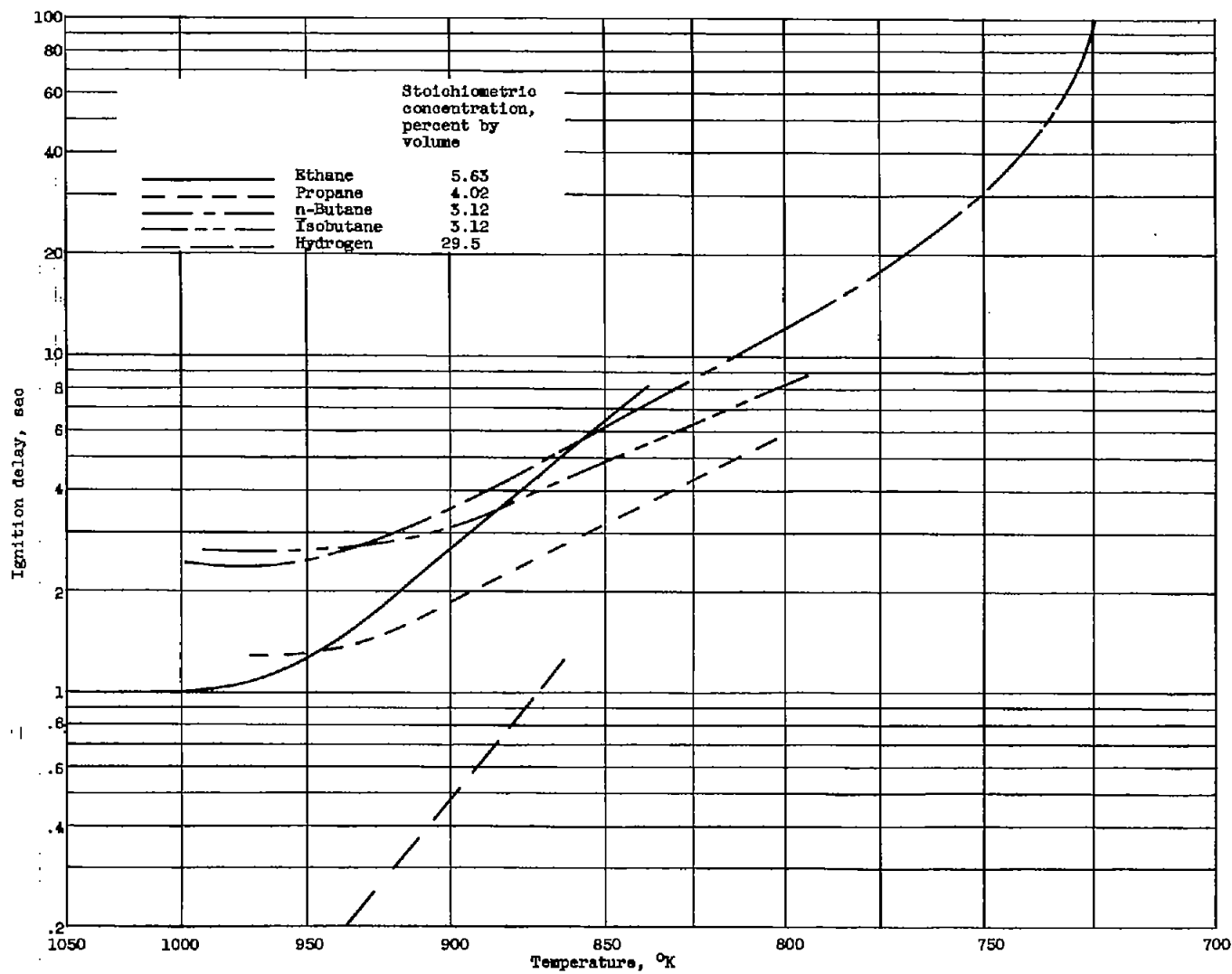
(b) Using fuel-air bypass and fuel bypass with large and small exhaust lines.

Figure 13. - Concluded. Variation of ignition delay with hydrogen concentration.



(a) Fuel concentration, 10 percent by volume.

Figure 14. - Variation of ignition delay with temperature for fuels studied.



(b) Fuel concentration, stoichiometric.

Figure 14. - Concluded. Variation of ignition delay with temperature for fuels studied.